

Pressure effects on superconducting properties of single-crystalline Co doped NaFeAs

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Resistivity and magnetic susceptibility measurements under external pressure were performed on single-crystals $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ ($x=0, 0.01, 0.028, 0.075, 0.109$). The maximum T_c enhanced by pressure in both underdoped and optimally doped $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ is the same, as high as 31 K. The overdoped sample with $x = 0.075$ also shows a positive pressure effect on T_c , and an enhancement of T_c by 13 K is achieved under pressure of 2.3 GPa. All the superconducting samples show large positive pressure coefficient on superconductivity, being different from $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$. However, the superconductivity cannot be induced by pressure in heavily overdoped non-superconducting $\text{NaFe}_{0.891}\text{Co}_{0.109}\text{As}$. These results provide evidence for that the electronic structure is much different between superconducting and heavily overdoped non-superconducting $\text{NaFe}_{1-x}\text{Co}_x\text{As}$, being consistent with the observation by angle-resolved photoemission spectroscopy.

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I. INTRODUCTION

Extensive experimental and theoretical efforts have been made to study the iron based superconductors since the discovery of superconductivity in F doped LaOFeAs .¹ Most of the parent compounds of the iron based superconductors undergo structural and spin density wave (SDW) transitions. With doping or applying high pressure, both the structural and SDW transitions are suppressed and superconductivity emerges. The so called "111"-type iron arsenide compound with the PbFCl structure, including LiFeAs ² and NaFeAs ³, has been regarded as a unique family which is superconducting without purposely doping or applying pressure. Although no long range antiferromagnetic order has been observed in LiFeAs ⁴, NaFeAs is reported to undergo three successive phase transitions at around 52, 41, and 23 K, which correspond to structural, magnetic, and superconducting transition, respectively.⁵ Although the resistivity of NaFeAs drops to zero at about 10K, it has been pointed out that the superconductivity is filamentary rather than a bulk phenomenon.^{6,7} With substitution of Co on Fe site, both magnetism and the structural distortion are suppressed, and bulk superconductivity with zero resistivity up to 20 K can be achieved.^{8,9} Full shielding fraction and large specific heat jump can be observed in single-crystalline optimally doped $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$ samples.⁷

Applying pressure has been proved to be an effective method to enhance the superconductivity transition temperature in many types of iron arsenide superconductors. It was revealed that the T_c of F-doped LaOFeAs was enhanced up to 43 K soon after the discovery of superconductivity in this system.¹⁰ In tetragonal FeSe , T_c increases from 8.5 K at ambient pressure to about 37 K

under $P = 8.9$ GPa, which is the largest pressure effect reported in iron based superconductors so far.¹¹ Pressure effects in electron doped 122-system $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ with different doping levels have been thoroughly studied. Applying pressure dramatically enhances T_c in the underdoped regime, whereas, the effect of pressure on T_c is rather small in the optimally doped and overdoped regimes.^{12,13} For the "111"-type Fe-pnictides, it is reported that the transition temperature of LiFeAs is suppressed linearly with pressure¹⁴, whereas, T_c of $\text{Na}_{1-x}\text{FeAs}$ polycrystal can be enhanced up to 31 K at about 3 GPa.¹⁵ This difference is attributed to the different ionic radius between Li and Na. However, in former high-pressure study the superconducting transition is rather broad due to the highly hygroscopic nature of polycrystalline NaFeAs sample. In order to study the intrinsic properties of this system, it is of great interest to investigate the combined effect of doping and pressure on superconducting properties of single-crystal samples. In this paper, we report the results of resistivity measurements under hydrostatic pressure for single-crystalline $\text{NaFe}_{1-x}\text{Co}_x\text{As}$, tracking T_c as a function of both pressure and doping level in different regions of the phase diagram. The initial slope of the pressure dependence of T_c , $(dT_c/dP)_{P=0}$, is positive in the whole superconducting doping regime of phase diagram. The value of pressure coefficient is comparably large among Fe-pnictides, even in the overdoped region. For the nonsuperconducting extremely overdoped sample, the pressure effect is negligible. T_c^{offset} as high as 31 K, generally consistent with the maximum T_c under pressure in polycrystalline NaFeAs , can be reached in both underdoped and optimally doped samples. The identical maximum T_c under pressure in different doping regions indicates that there is a universal maximum transition temperature of about 31 K in electron-doped NaFeAs , which can be obtained by applying high pressure or combined effect of pressure and doping.

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II. EXPERIMENTAL DETAILS

High-quality $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ single crystals were grown by the conventional high temperature solution growth method using the NaAs self-flux technique. Details of the growth procedures were provided in our previous work.⁷ Electrical resistivity was measured using the ac four-probe method. Pressure was generated using a Be-Cu pressure cell with a Teflon cup which was filled with Daphne Oil 7373. The pressure applied in the resistivity measurement was determined by shift of the superconducting transition temperature of pure Sn.¹⁶ The magnetic susceptibility was measured under pressure up to 6.1 GPa in a diamond anvil cell (DAC). The pressure transmitting medium was Daphne Oil 7373 and the pressure was measured at room temperature by ruby fluorescence spectroscopy. The resistivity measurements were performed using a Quantum Design physical properties measurement system (PPMS-9), and the magnetic susceptibility was measured using a superconducting quantum-interference device magnetometer (SQUID-MPMS-7T, *Quantum Design*).

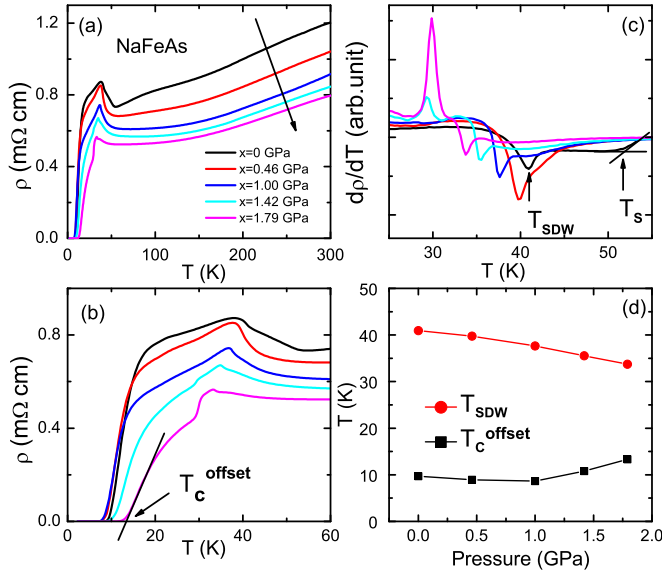


FIG. 1: (color online). (a): Temperature dependence of in plane resistivity for NaFeAs under different pressures. Arrow indicates the direction of the increasing pressure. (b): Expanded plot of the temperature dependence of resistivity under various pressure around T_c . (c): The derivative of the in-plane resistivity $d\rho/dT$ and the criteria by which we infer T_s and T_{SDW} . (d): T_c and T_{SDW} as a function of pressure for the parent compound NaFeAs.

III. RESULTS AND DISCUSSION

Fig. 1(a) shows the temperature dependence of resistivity for NaFeAs under different pressures. Two anomalies in the resistivity curve are observed at 51 K and

41 K under ambient pressure, which are consistent with previous reports.⁵ These anomalies have been proved by neutron scattering experiment to arise from the structural and SDW transition, respectively.⁶ With increasing pressure the anomalies corresponding to the SDW transition is gradually suppressed to lower temperature, whereas the structural transition quickly becomes undetectable. The suppression of the resistive anomalies can also be seen in the derivative of resistivity shown in Fig.1(c). We use the same criteria to infer the structural and SDW transitions from the resistivity as described in Ref.17, which has been confirmed by specific heat and magnetic susceptibility measurements.⁷ As shown in Fig.1(b), the superconducting transition is rather broad, and the three transitions take place in a narrow temperature range, thus it is difficult to define the T_c^{onset} . We use the criterion T_c^{offset} to describe the superconducting transition temperature in this paper, the definition of which is shown in Fig.1(b). With the applied pressure increasing, T_c^{offset} firstly decreases slightly, then increases quickly with pressure higher than 1 GPa. The highest T_c^{offset} we can achieve is 11.9 K at $P = 1.79$ GPa. The data of NaFeAs under pressure are summarized in Fig.1(d). The phase diagram $T(P)$ is similar to that of underdoped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ ¹², in which T_{SDW} is suppressed gradually while superconductivity is enhanced by applied pressure.

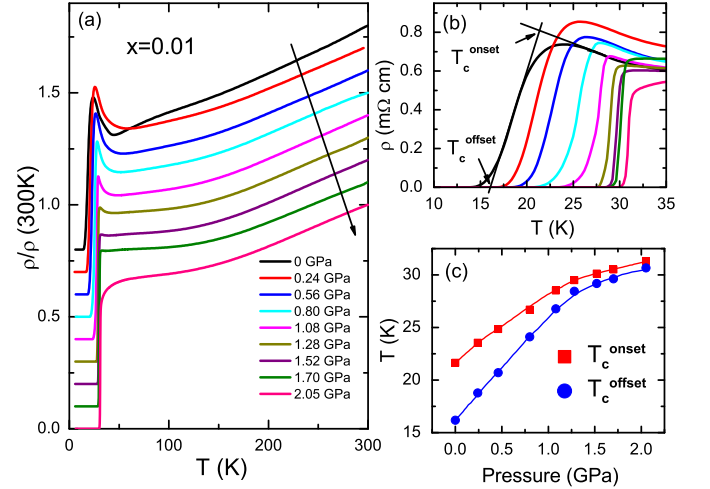


FIG. 2: (color online). (a): Temperature dependence of the resistivity under different pressures up to 2.05 GPa for $\text{NaFe}_{0.99}\text{Co}_{0.01}\text{As}$, normalized to the room temperature value. Each subsequent data set is shifted downward by 0.1 for clarity. (b): Enlargement of the low temperature resistivity and the criteria used to determine the onset and offset temperatures for the superconducting transitions. (c): T_c as a function of pressure for the underdoped crystal $\text{NaFe}_{0.99}\text{Co}_{0.01}\text{As}$.

For the underdoped sample $\text{NaFe}_{0.99}\text{Co}_{0.01}\text{As}$ with $T_c^{\text{offset}} \sim 16\text{K}$, the kinks in resistivity curves associate with the structural and SDW transition are distinct at ambient pressure. Once the external pressure is applied,

the kinks quickly become obscure and then indistinguishable, similar to the case in doped 122-system.¹² As shown in Fig.2(a), the low-temperature resistive upturn corresponding to the structural and/or magnetic transition are progressively suppressed, and ultimately vanished at $P = 2.05$ GPa, at which the highest superconducting transition temperature about 30.7 K is obtained. The criteria used to determine the onset and offset temperature of superconducting transition are shown in Fig 2(b). Since the onset temperature of superconductivity is ambiguous in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$, we write T_c for T_c^{offset} for convenience hereafter. As reported in underdoped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, the critical pressure at which the high temperature transition disappears coincides rather well with the pressure at which T_c is highest and the superconducting transition is narrowest.¹³ The pressure coefficient dT_c/dP is 9.6 K/GPa below 1.28 GPa, and the pressure coefficient between ambient and the pressure at which T_c reaches its maximum is 7.06 K/GPa, even larger than the pressure coefficient of FeSe (3.2 K GPa^{-1}).¹¹ The pressure effect coefficient based on the T_c^{onset} is about 4.7 K/GPa which is still relatively large in iron pnictides. In the phase diagram shown in figure 2(d), it is obvious that the superconducting transition width become narrower with increasing the pressure. The sharp superconducting transition observed at 2.05 GPa indicates that the pressure condition is still hydrostatic.

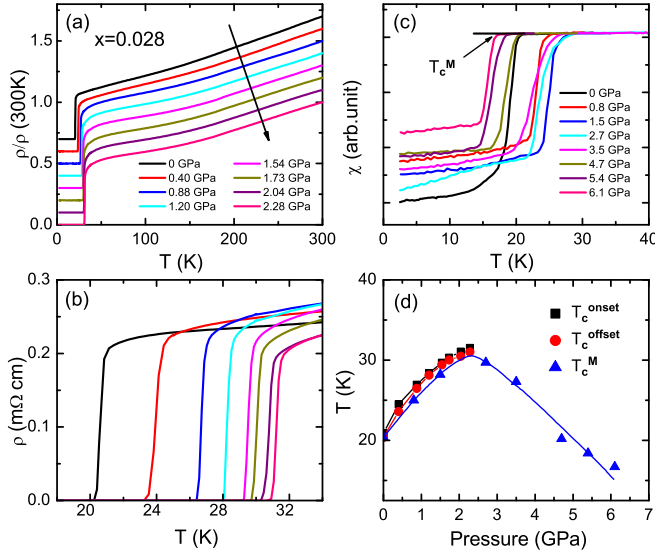


FIG. 3: (color online). (a) Temperature dependence of in plane resistivity for $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$ under various pressures. Data are shown normalized by room temperature resistivity, successive data sets are offset vertically by 0.1 for clarity. (b) The data in panel (a) is plotted in low temperature range for clarity. (c): The plot of Magnetic susceptibility as a function of temperature in zero-field cooled measurements up to 6.1 GPa. (d): Evolution of T_c determined by resistivity and susceptibility measurements with the applied pressure.

For the optimally doped sample $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$,

the anomalies associated with the structural or SDW transition are suppressed completely by Co doping. As shown in Fig.3(a), T_c measured by in-plane resistivity increases monotonously from 20.4 K to 31.0 K with increasing the applied pressure from zero to 2.28 GPa. The pressure coefficient is 4.67 K/GPa, much larger than 1 K/GPa in optimally doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ ¹² and comparable with 5 K/GPa in optimally doped $\text{LaFeAsO}_{1-x}\text{F}_x$.¹⁰ In order to establish the complete superconducting dome in the phase diagram. We carried out the magnetic susceptibility measurement using the diamond anvil cell (DAC) technology. Pressures up to 6.1 GPa were applied on $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$ single crystal. The T_c^M values under various pressures are determined from the beginning of deviation from the extrapolated line of the normal state M - T curve as shown in Fig.3(c). Fig.3(d) displays the $T(P)$ phase diagram based on the resistivity and magnetic susceptibility measurements. The T_c^M measured by DAC technology initially increases monotonously, and begins to decrease when pressure is higher than 2.3 GPa. The behavior of transition temperature obtained by resistivity and magnetic susceptibility are highly consistent with each other. The highest transition temperature obtained by our measurement is 31.0 K, where the transition width is 0.5 K which is considerably sharp. The $T_c = 31.0$ K presented here is the highest in the 111 system up to now.

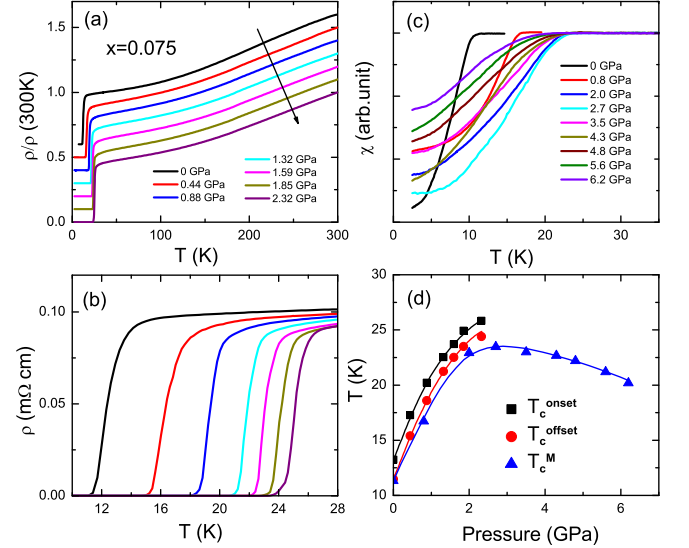


FIG. 4: (color online). (a): Temperature dependence of in plane resistivity for $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ under various pressures, Successive data sets are offset vertically by 0.1 for clarity. (b): The same data shown in (a) around the superconducting transition. (c): Temperature dependence of magnetic susceptibility under different pressures for $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$. (d): The T_c obtained from resistivity and susceptibility measurements as a function of pressure.

Fig.4(a) and Fig.4(c) display the pressure dependence of the in-plane resistivity and magnetic susceptibility

of overdoped sample $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$, respectively. The superconducting transition temperature of this overdoped sample is 11.5 K at ambient pressure. Similar to the case in optimally doped $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$, the T_c increases monotonously up to 24.5 K with increasing the applied pressure to 2.32 GPa. Domelike shape of $T_c(P)$ was revealed by magnetic susceptibility measurement, from which we can infer that the highest superconducting transition temperature in $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ is about 24.5 K with the uncertainty less than 1 K. A large enhancement of T_c by 13 K, which is comparable to those in underdoped and optimally doped samples, is still exist in this overdoped composition. The pressure coefficient of $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ is 5.57 K/GPa, even higher than that of the optimally doped sample. The large pressure coefficient of overdoped $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ is obviously different from those in other iron-pnictide superconductors, which are rather small or change their sign from positive to negative in the overdoped regime in the phase diagram.^{12,18–20}

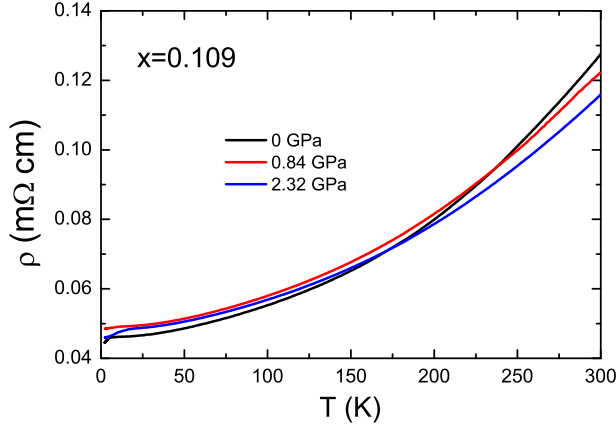


FIG. 5: (color online). Temperature dependence of in plane resistivity for heavily overdoped non-superconducting crystal $\text{NaFe}_{0.891}\text{Co}_{0.109}\text{As}$ under various pressures.

When external pressure was applied on the extremely overdoped sample $\text{NaFe}_{0.891}\text{Co}_{0.109}\text{As}$ which shows no superconductivity down to 2 K at ambient pressure, we cannot observe the pressure-induced superconductivity with applied pressure up to 2.32 GPa. Besides, both the magnitude and the behavior of the resistivity do not change much with the applied pressure.

The effect of applied pressure on the superconducting transition temperature of $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ is illustrated in Fig.6(a), where the maximum T_c under pressure and T_c at ambient pressure are plotted as a function of doping level x . Because the maximum transition temperature has not been reached in undoped NaFeAs , we use the maximum value reported by Zhang *et al.* for this composition.¹⁵ Large positive pressure coefficient dT_c/dP is observed in all the superconducting compositions, even in the overdoped regime. The maximum tran-

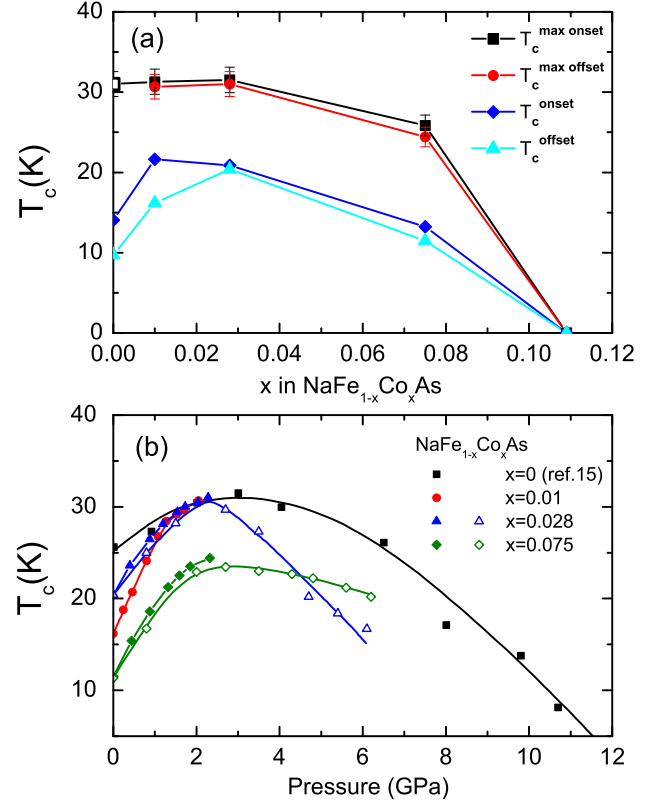


FIG. 6: (color online). (a): Comparison of T_c at ambient pressure and the maximum T_c achieved under applied pressure at various Co concentrations. The open square represent the T_c^{onset} reported by Zhang *et al.*¹⁵ (b): $T(P)$ phase diagram of $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ with different doping levels. Open and filled symbols represent data obtained from susceptibility and resistivity measurements, respectively.

sition temperatures in NaFeAs , $\text{NaFe}_{0.99}\text{Co}_{0.01}\text{As}$ and $\text{NaFe}_{0.972}\text{Co}_{0.028}\text{As}$ are all around 31 K. $T(P)$ phase diagram of $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ system for various x is shown in Fig.6(b). It is obvious that the maximum transition temperature in undoped, underdoped and optimally doped sample is strikingly the same. These results indicate that there is a universal maximum transition temperature in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ of about 31 K, which can be achieved by applying a critical pressure of $P = 2-3$ GPa.

For the undoped and underdoped samples, applied external pressure suppresses the structural and SDW transitions, and enhances superconductivity simultaneously. This behavior is similar to the pressure effect in $\text{LaFeAsO}_{1-x}\text{F}_x$ ¹⁰ and 122-systems.^{12,19,21,22} Although the T_c and normal state resistivity behavior evolve systematically with Co doping, the maximum transition temperature enhancement and corresponding critical pressure is nearly the same in all the superconducting samples. These properties are different from the case in most of the iron based superconductors that pressure effect is different in different regions of electronic phase diagram. An identical maximum T_c of 31 K un-

der doping as well as pressure has also been reported in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$.²³ In the P doped Ba-122 system, phosphorous substitution could be regarded as chemical pressure, which changes Fe-Pn distance and causes similar effects on superconductivity to the physical pressure. Though the Co substitution in NaFeAs is referred to as electron doping, different from the replacement of As by P which is referred to as isovalent substitution, it is likely that the pressure-induced enhancement of T_c in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ is also associated with the optimization of the structural parameters of FeAs layers, including the As-Fe-As bond angle and anion height.²⁴ One possible reason for lower maximum transition temperature obtained in overdoped $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ single crystal is that the superconductivity is disturbed by the disorder or additional scattering induced by excess cobalt doping. This phenomenon is different from the case of overdoped $\text{LaFeAsO}_{1-x}\text{F}_x$, in which the conducting layer is not affected by F doping and the highest transition temperature acquired in optimal and overdoped samples are almost the same.¹⁰

The overdoped superconducting sample $\text{NaFe}_{0.925}\text{Co}_{0.075}\text{As}$ still has considerable positive pressure coefficient which is rare in Fe-pnictide superconductors. However, when the pressure is applied on the extremely overdoped non-superconducting sample, no superconductivity induced by pressure can be observed. It has also been reported that temperature linear dependent susceptibility can be observed in high temperatures for all the superconducting samples and the breakdown of the temperature linear dependent susceptibility in the overdoped region coinciding with the disappearance of superconductivity.⁷ These phenomena indicate that there is a sudden change in the electronic structure between the superconducting compositions and the heavily overdoped non-superconducting phase. This conclusion is supported by the experimental results of STM investigations²⁵ and the angle-resolved photoemission spectroscopy (ARPES) studies.²⁶ The STM study revealed that the high energy dI/dV spectra of superconducting $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ remain nearly the same, whereas, the high energy spectrum suddenly started to shift to the lower energy substantially for the sample with $x = 0.109$. The direct measurements of the electronic structure of $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ by ARPES revealed that all the superconducting $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ compounds have similar band structures and small relative Fermi level shifts. However, the $x = 0.109$

compound in the heavily overdoped regime shows a large Fermi level shifts (about 100 meV) relative to the optimally doped compounds, and its band structure is significantly changed as the hole-like bands around the zone center disappears and an electron pocket appears instead, which means the consequent Fermi surface consists of electron pockets only in this sample. The drastic change in electronic structure for the heavily overdoped nonsuperconducting samples could explain the observed properties of pressure effect.

IV. SUMMARY

In conclusion, we have performed resistivity and magnetic susceptibility measurements under various pressure on $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ ($x=0, 0.01, 0.028, 0.075, 0.109$) single crystals. In the undoped and underdoped compounds, structural and SDW transitions are gradually suppressed while superconductivity is enhanced by applied external pressure. A universal maximum transition temperature of about 31 K under external pressure is observed in underdoped and optimally doped $\text{NaFe}_{1-x}\text{Co}_x\text{As}$. The superconducting transition temperature of $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ is strongly enhanced in the whole superconducting regime of the phase diagram, and the pressure effect is considerably large compared to other iron pnictides. The large positive pressure coefficient in the optimally and overdoped region is different from that in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, and disappears simultaneously with the superconductivity in the phase diagram. These results could be explained as originated from the similarity of electron structures within the superconducting dome, and a drastic change of the electron structures between the superconducting overdoped regime and the non-superconducting heavily overdoped regime, which correspond to the conclusions of STM and ARPES measurements.

Acknowledgements

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